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Chromium redox transformations in diffusion-limited domains of soils: Linking micro- and macro-scale processes

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The fate of redox-sensitive contaminants in soils is complex because of the broad range of transport times and reaction rates encountered over short distances. Within soil aggregates, reactive transport processes are commonly only inferred. Direct measurements of chromium transport and redox transformations within soil aggregates were obtained using micro-XANES mapping and spatially-resolved microbial community analyses. During the diffusion-limited contamination process, more Cr(VI) was transported, but to shorter distances, in more microbially active aggregates. Sharply terminated diffusion fronts, within 2 to 10 mm of the aggregate surface, result from increasing Cr(VI) reduction rates with depth. Infusion of organic carbon into previously Cr(VI)-contaminated aggregates promoted reduction to Cr(III). Reoxidation of Cr(III) over longer times is occurring in the lower organic carbon soils. These results show that intra-aggregate Cr dynamics are strongly diffusion-limited in more microbially active systems, that bulk soil chemical and microbial characterization can obscure relevant biogeochemical processes, and that the long-term stability of reduced Cr needs to be understood.